Title

Appropriate NaI (Tl) Detector for Measuring Gamma-Ray Emitted from ^{137}Cs in Sea Water Using Monte Carlo Code MCNP.4C Simulation

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Abstract

The systems most commonly used for low level gamma radiation spectroscopy in seawater, are based on NaI (Tl) detectors, which are characterized by high-detection efficiency and low cost. ^{137}Cs is the most important radioactive contaminant, produced from the atmospheric bomb tests, discharges from nuclear reprocessing plants, leaking of concealed nuclear waste and in general of the sea (ocean). To select gamma ray of radiocaesium counting detector in the sea water near Bushehr nuclear power plant (B.N.P.P), MCNP.4C were used to simulate three different dimensions of (2in×2in (long ×diameter), 3in×3in and 6in ×3in) cylindrical NaI (Tl) detectors and in turn the volume of sea water, and chemical and physical characteristics of the sea water near the B.N.P.P and optimum fluxes was calculated. Response of detectors to gamma radiations by using Monte Carlo code MCNP.4C for simulated condition was calculated. The obtained results showed that NaI (Tl) detector with 6in ×3in dimension is a suitable detector for online radiocaesium monitoring in Bushehr sea water.

Key words

Simulation; NaI Detector; Gamma radiation spectroscopy; Cs-137; Monte Carlo code MCNP.4C

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Introduction

Today, Development of a system for measuring radioactivity in the water environment is an important scientific priority for the marine sciences and especially for the operational oceanography [1].The systems most commonly used for gamma ray spectrometry are based on NaI (Tl) or HPGe detectors. HPGe-based systems have the advantage of good energy resolution and hence excellent radionuclide identification capability. The advantage of NaI (Tl)-based systems is primarily related to the high detection efficiency of NaI (Tl) crystals at much lower cost than equivalent Ge crystals [1,2].

The γ-radiation level is not constant. It fluctuates time to time due to many reasons, which could not be easily identified by the gross count measurements since the information is far less than the high-resolution spectrometric data. It is important to understand the phenomenon of monitoring level fluctuation due to natural causes, since the purpose of the monitoring is to find artificial contributions to the radiation levels. The construction of optimal systems for describing and quantifying the radioactive contamination distribution in a scientific and cost-effective manner is one of the very important demands in the scientific radio ecological community. Many new monitoring tools and methods are tested in the few last years [3-6].

NaI (Tl) detectors have been widely used since their discovery by Hofstadter in 1948. Their response to Gamma-rays are important especially for the online elemental analysis [7]. NaI (Tl) is recommended for all nuclide identification, because of high efficiency being rugged to thermal and mechanical shock, relatively low cost, and availability in wide variety of sizes. The light output of NaI (Tl) results in a small contribution due to photoelectron statistics in the measured resolution. The artificial radionuclide is the greatest radiological significance in the marine environment [7-9]. ^{137}Cs is of special interest among the artificial radionuclides, because it is a long-lived radionuclide (30.05 years); it is used as a radionuclide tracer in seawater and constitutes the artificial radionuclide of greatest 3 radiological significance in the marine environment [10-12].

MCNP Monte-Carlo code is a general purpose computer code which is particularly useful for phenomena which are random in nature such as interactions of nuclear particles with materials. It does not solve the transport equation for the neutron, but rather simulates one neutron at a time and records its history. MCNP can be used for simulation of neutron, electron, photon and/or a combined neutron/electron/photon transport problem [13].

Methods &Materials

Simulation

In the present work, the MCNP.4C code has been implemented as the Monte Carlo simulation package of choice. The code simulates all relevant physical processes taking place in matter, along the passage of elementary particles from the source to the detector of any configuration. A detailed description of the geometry of the experimental setup and environment with respect to their dimensions, materials and shapes, as well as the particle generator, is required by the program to simulate and store the data of each particle from its generation to full deposition of its energy in the detector. Monte carlo MCNP.4C code was used to simulate a favorite condition close to real state of sea near the B.N.P.P .The geometry of detectors with 2 in×2 in (long × diameter), 3 in×3 in, and 6 in ×3 in cylindrical dimensions NaI (Tl) with Al enclosure of detectors in 1 mm thickness, and other detector accessories consisted of PMT, electronic device, etc. According to real size and material are assumed in simulation. In this simulation, other conditions such as the radionuclide sources strength, related atomic and mass numbers, half life and decay of existing radionucleas and emitted particles, chemical and physical characteristic of sea water are also defined according to the real conditions of sea water. Density of water is calculated as a mixture density using the following formula:

$$\rho_{mix}=W_1/\rho_1+\ldots+Wn/\rho_n \quad (1)$$

where ρ_{mix} is density of mixture; W_1 and Wn are weight fractions of existing element in sea water and ρ_1 and ρ_n are relevant densities of each element.

MCNP.4C code and Monte Carlo data analysis

In the Monte Carlo simulation, the relevant physical quantity is the energy deposited into the detector crystal. The results obtained by non analog simulation are biased by the variance reduction technique followed by a correction. A particle with certain weight w is introduced and calculated for each particle record. If an event occurs, the weight w is added to the corresponding energy in the histogram rather than incrementing by one unit. In order to compare calculated and experimental efficiencies, it is necessary to simulate the statistical fluctuations in the process of charge carrier production and pulse electronic analysis. For this purpose, a Gaussian distribution with parameters extracted from experimental data is applied to the deposited energy [12].

Analog Monte Carlo methods are based in determining efficiency by simulating all relevant physical processes taking place along the path of a photon emitted by the source. The history of each individual primary particle consists of its emission by the source, interaction with the detector and surrounding materials, production as well as transport of secondary particles, and track until the photon escapes or undergoes a photoelectric interaction in the crystal, depositing all of its energy.

Since no approximations are needed, there is no limitation on the source–detector configuration. The main disadvantage of these analog Monte Carlo calculations is that a large number of records (> 10^5–10^6 primary photons) must be simulated to obtain a statistical uncertainty of less than 1%. Therefore, analog Monte Carlo methods need long computing times. Thus, it is desirable to introduce a variance reduction technique to improve its computational efficiency (non-analog Monte Carlo method) [14]. The present research introduces a simple variance reduction scheme based on directional bias. The goal of this method is to simulate only primary photons that are emitted from the sample in directions towards the detector active volume.

Results and discussion

Monte carol MCNP.4C code was used to simulate a favorite condition close to real state of the sea near B.N.P.P .The geometries of detectors are 2 in×2 in (long ×diameter), 3 in×3 in, and 6 in ×3 in dimension with Al enclosure of detectors in 1 mm thickness, and other detector accessories consists of PMT, electronic device, etc. All concentration of existing ions and elements in sea water are studied and data which is used in the simulation extracted from IAEA annual reports. Three dimensions of

NaI (Tl) detector is selected and all characteristics of these crystals are investigated and used as real properties in MCNP.4C simulation program. To avoid any turbulency caused by water Debby effecting detectors, crystal of detector damaging and achieve steady output and a watertight cylindrical aluminum enclosure have been simulated, which houses the above-mentioned NaI system together with the appropriate electronics. The aluminum enclosure in 1mm thickness is considered. Figure 1 shows the schematic diagram of simulated detectors. Detectors assumed immersed in the middle of simulated sea water with defined volume and surrounded with water homogenously. Radiocaesium point is assumed in homogeny propagating. The volume of sea water is assumed to be infinite in physical scale.

Twenty-two different volumes (50, 100, 150, 200, 250, 300, 350, 400, 450, 500, 550, 600, 650, 700, 750, 800, 850, 900, 950, 1000, 1050, 1100 liters) are considered that are increased in 50 liter interral. Each simulated system is studied separately and to avoid systematic error, each calculation is repeated for 5 times. .Accumulated gamma radiation of ^{137}Cs with NaI (TL) detectors is calculated with MCNP.4C code. Table 1. shows calculated fluxes from MCNP.4C for simulated system with 2 in×2 in, 3 in×3 in and 6 in ×3 in NaI Tl) detectors. Standard deviation (STD) of resulted flux for each simulated system is calculated. Relevant STD and RSD for resulted fluxes of system with 2 in×2 in, 3 in×3 in and 6 in ×3 in, NaI (Tl) detectors is given in Table 2.

Refer to Table 1, Flux1 is calculated for system with 2 in×2 in, flux2. is calculated for system with 3 in×3, in and flux3 is calculated for system with 6 in×3 in, NaI (Tl) detectors in count/l.sec. Related

Table 1- Calculated fluxes for simulated system with 2in×2 in, 3 in×3 in and 6in ×3in, NaI (Tl) detector.

System No.	Volume (lit)	flux.1 (count/lit)	flux.2 (count/lit)	flux.3 (count/lit)
1	50	2.36×10^{-2}	7.16×10^{-2}	1.43×10^{-1}
2	100	6.79×10^{-2}	1.32×10^{-1}	2.68×10^{-1}
3	150	4.48×10^{-2}	2.02×10^{-1}	3.97×10^{-1}
4	200	6.42×10^{-2}	2.02×10^{-1}	$4.25\times10^{-}$
5	250	8.67×10^{-2}	2.06×10^{-1}	5.38×10^{-1}
6	300	1.08×10^{-1}	2.68×10^{-1}	5.56×10^{-1}
7	350	8.37×10^{-1}	2.77×10^{-1}	6.75×10^{-1}
8	400	1.07×10^{-1}	3.20×10^{-1}	7.08×10^{-1}
9	450	1.13×10^{-1}	3.79×10^{-1}	7.90×10^{-1}
10	500	1.31×10^{-1}	3.90×10^{-1}	7.98×10^{-1}
11	550	1.38×10^{-1}	3.99×10^{-1}	7.99×10^{-1}
12	600	1.38×10^{-1}	4.54×10^{-1}	7.99×10^{-1}
13	650	1.41×10^{-1}	4.54×10^{-1}	7.99×10^{-1}
14	700	1.43×10^{-1}	4.56×10^{-1}	7.99×10^{-1}
15	750	1.43×10^{-1}	4.56×10^{-1}	7.99×10^{-1}
16	800	1.43×10^{-1}	4.56×10^{-1}	7.99×10^{-1}
17	850	1.44×10^{-1}	4.56×10^{-1}	7.99×10^{-1}
18	900	1.44×10^{-1}	4.57×10^{-1}	8.00×10^{-1}
19	950	1.44×10^{-1}	4.57×10^{-1}	8.00×10^{-1}
20	1000	1.44×10^{-1}	4.57×10^{-1}	8.00×10^{-1}
21	1050	1.44×10^{-1}	4.57×10^{-1}	8.00×10^{-1}
22	1100	1.44×10^{-1}	4.57×10^{-1}	8.00×10^{-1}

Table 2- Standard deviation of calculated fluxes, RSD and, mean of calculated fluxes for simulated system.

Detector dimension	STD	Mean of fluxes (count/lit)	RSD
2in×2in	0.037	1.81e-01	3.15e+01
3in×3in	0.125	3.58e-01	3.51e+01
6in ×3in	0.199	6.77e-01	2.95e+0

time of flux calculation using MCNP.4C software for all simulated system is identical and with all NaI detector it is between 0.06-0.08 minutes. Maximum flux of radiocaesium is calculated for system of 6in×3in NaI (Tl) detector and minimum flux is calculated for system of 2 in×2 in NaI (Tl) detector. Maximum STD is resulted for the system with 6 in×3 in NaI (Tl) detector, and minimum STD deviation is resulted for the system with 2in×2 in NaI (Tl) detector. Calculated RSD for simulated system with 6 in×3 in NaI (Tl) detector is minimum, so there is a better precision of calculated flux in this system. Diagram of calculated flux vs. simulated volume is given in Figures 2, 3, and 4 for simulated system with 2 in × 2 in, 3 in × 3 in and 6 in ×3 in NaI (Tl) detectors, respectively. The diagrams show, that increase of flux resulted in the increase of volume. In system with 2in×2 in NaI (Tl) detector, minimum flux is 2.36×e-02 count/l.sec and maximum flux is 1.44e-01count/l.sec, regarding background of radiocaesium in the sea near the B.N.P.P (extracted from IAEA annual reports) $1.81\times10^{-3}\pm0.36$ Bq/L, and limit of detection (LOQ) in selected detector is 0.2-0.3 Bq/l, so this detector is not suitable for online radiocaesium monitoring in Bushehr sea water. In similar way, minimum calculated flux for system with 3in×3in NaI (Tl) detector is 7.16e-02count/l.sec and maximum flux is 4.57e-01count/l.s, and in simulated system with 6 in×3 in NaI (Tl) detector, maximum flux is 8.00e-01 count/l.s which is two times larger than calculated maximum flux in system with 3 in×3 in NaI (Tl) detector.

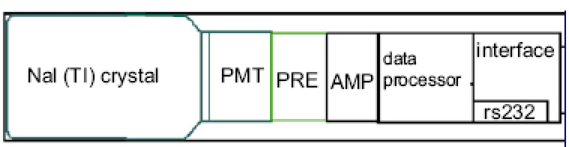

Figure1- The schematic diagram of simulated detectors.

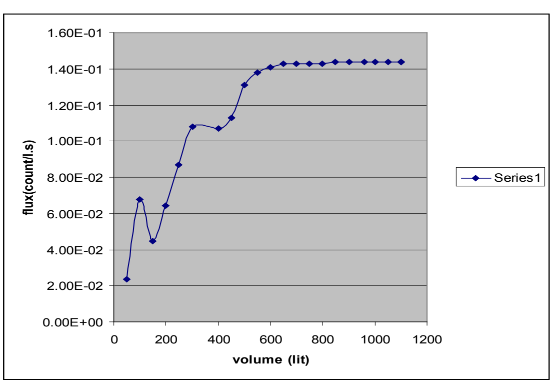

Figure 2- Variation of calculated flux vs. volume in the system with 2 in×2 in NaI (Tl) detector.

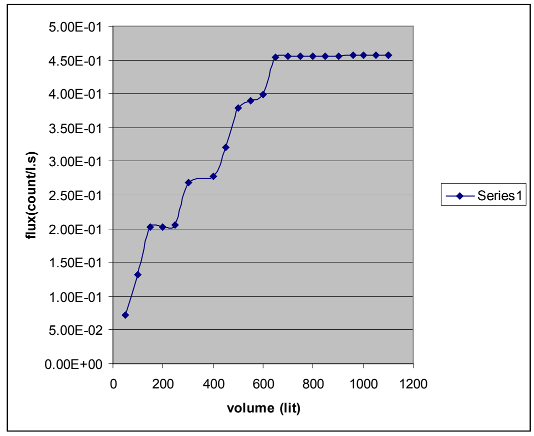

Figure 3- Variation of calculated flux vs. volume in the system with 3 in×3 in NaI (Tl) detector.

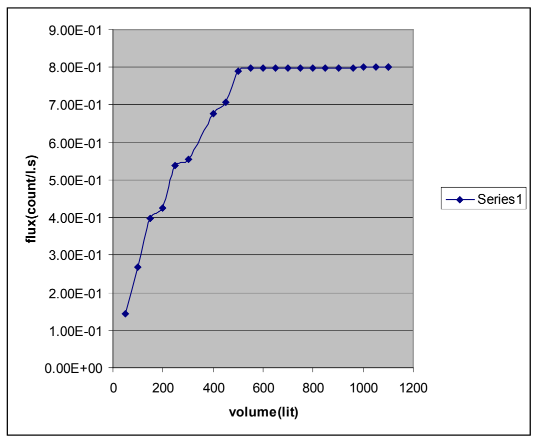

Figure 4- Variation of calculated flux vs. volume in the system with 6in×3in NaI (Tl) detector.

In the simulated system with 3 in×3 in NaI (Tl) detector up to 650 liter, flux increases but beyond this volume, flux is nearly constant, so volume of 650 liter is defined as a physical infinite volume. In system with 6 in×3 in NaI (Tl) detector, infinite volume is 500 liter which is less than that of simulated system with 3 in×3 in NaI (Tl) detector. Comparison of these results indicates that the flux of 6in×3in has a better condition such as low RSD and maximum calculated fluxes for using online measuring ^{137}Cs in sea water. Therefore, it is concluded that for online radiocaesium monitoring in sea near the B.N.P.P, 6in×3 in NaI (Tl) detector is a more suitable.

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